

Electronic Supplementary Information (ESI)

Synthesis and characterization of **1** and **2**.

[Mn^{II}(Ni^{II}L)₂]**·**2CH₃OH (**1**): Mn^{II}Cl₂·4H₂O (0.02 g, 0.1 mmol) in methanol (30 cm³) and Et₃N (0.02 g, 0.2 mmol) in methanol (20 cm³) were added to a methanol solution (50 cm³) of [Ni^{II}(HL)] (0.097 g, 0.2 mmol). The mixture was left at room temperature for three days to form orange crystals. Yield: 0.052 g (48%). Anal.: calcd for C₅₄H₅₆N₆Ni₂MnO₈ = [Mn^{II}(Ni^{II}L)₂]**·**2CH₃OH: C 59.54, H 5.18, N 7.71, Ni 10.78, Mn 5.04%; found: C 59.37, H 4.30, N 7.74, Ni 10.78, Mn 5.09%. IR (KBr): ν (C=N) 1628 cm⁻¹. Λ_M : 2.01 S mol⁻¹ cm² in DMF (ca. 0.4 mM). UV-Vis (DMSO): 555 (log ϵ/M^{-1} cm⁻¹: 59) and 368 nm (4.41).

[Fe^{III}(Ni^{II}L)₂](NO₃)**·**C₂H₅OH (**2**): Fe^{III}(NO₃)₃·9H₂O (0.04 g, 0.1 mmol) in ethanol (10 cm³) and Et₃N (0.02 g, 0.02 mmol) in ethanol (10 cm³) were added to an ethanol solution (40 cm³) of [Ni^{II}(HL)] (0.097 g, 0.2 mmol). The mixture was left in a refrigerator for two weeks to form dark purple crystals. Yield: 0.043 g (38%). Anal.: calcd for C₅₄H₅₄FeN₇Ni₂O₁₀ = [Fe^{III}(Ni^{II}L)₂](NO₃)**·**C₂H₅OH: C 57.18, H 4.80, N 8.64, Ni 10.35%; found: C 57.04, H 3.90, N 8.73, Ni 10.14%. IR (KBr): ν (C=N) 1631; ν (NO₂) 1278 cm⁻¹. Λ_M : 55 S mol⁻¹ cm² in DMF (ca. 0.4 mM). UV-Vis (DMSO): 514 nm (log ϵ/M^{-1} cm⁻¹: 3.64) and 344 nm (4.46).

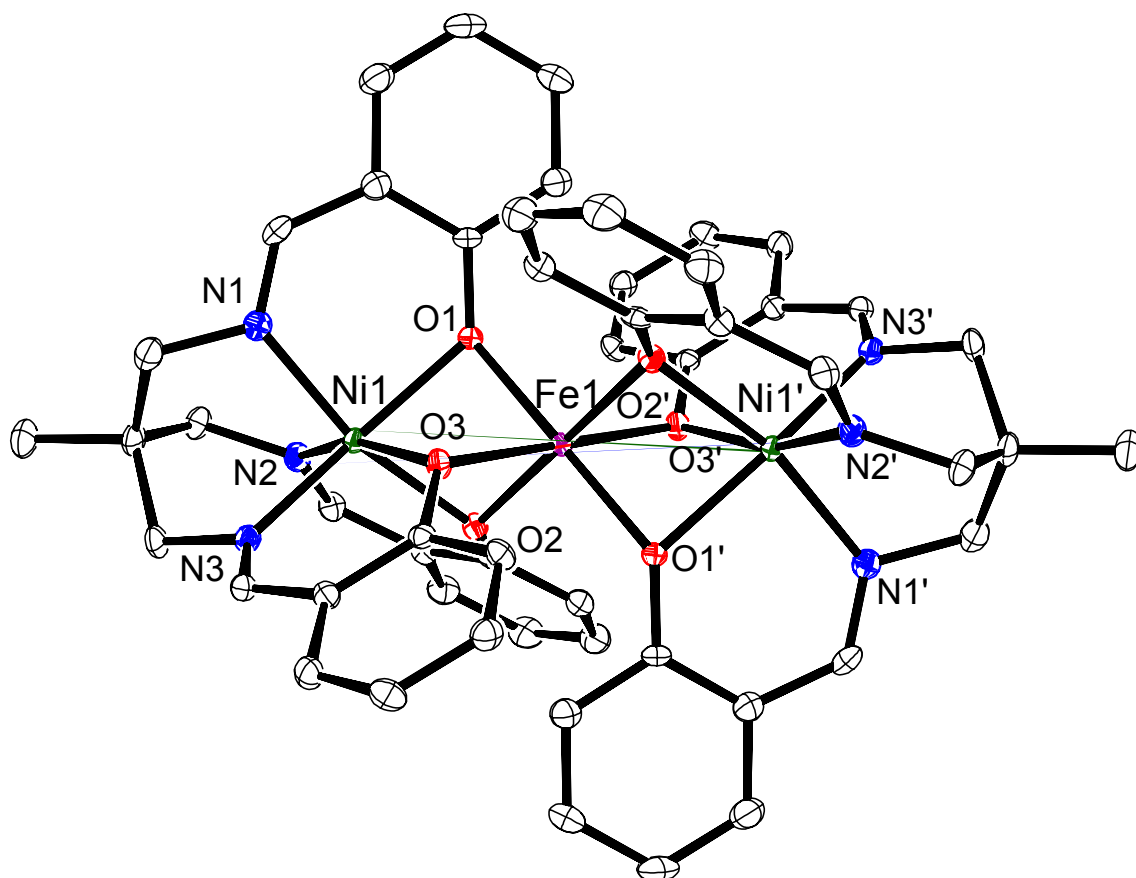


Fig. S1. X-ray molecular structure of 2.

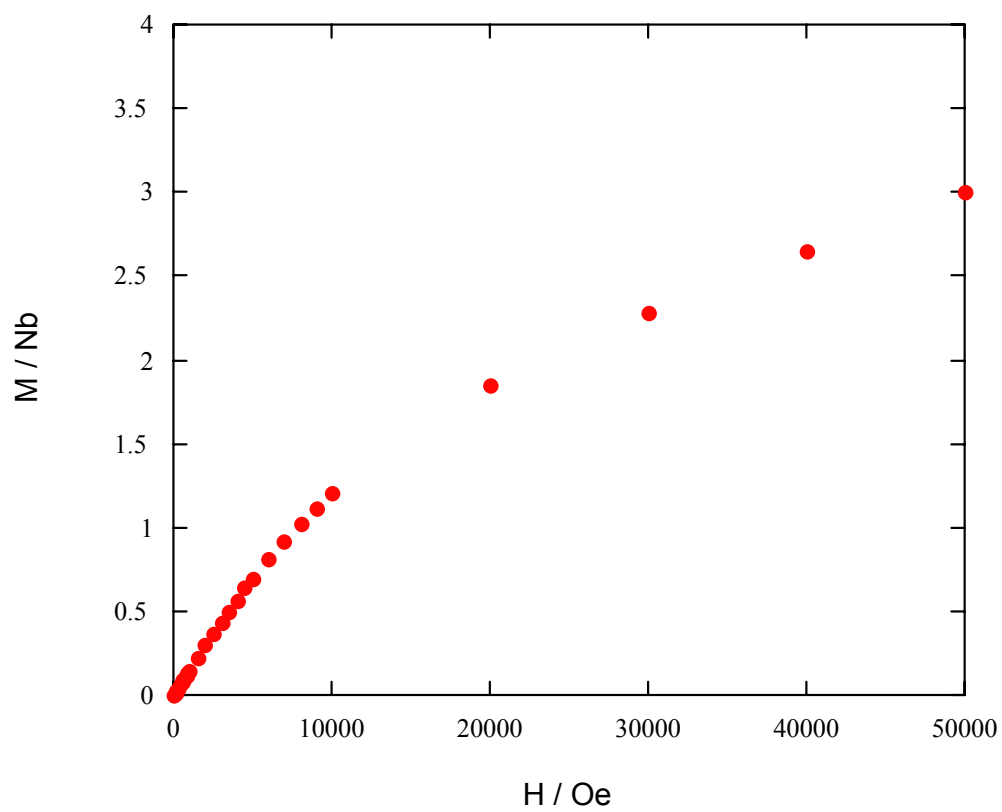


Fig. S2. Field dependence of magnetization at 1.9 K for **2**.